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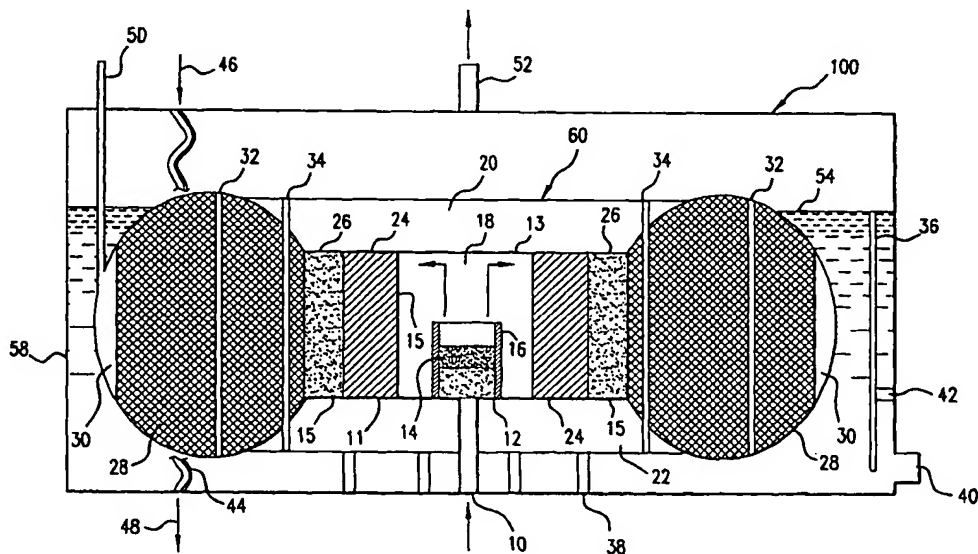
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(54) Title: APPARATUS FOR PRODUCING HYDROGEN



(57) Abstract: A fuel processor for converting a hydrocarbon or an oxygenate feed stream to produce a fuel stream for a fuel cell is provided wherein the heat required to sustain the reforming reaction and the cooling required to moderate and direct high and low-temperature shift reactions occur in a thermosiphon boiler vessel (100). The thermosiphon boiler vessel (100) provides an essentially isothermal steam generator and heat removal system which improves the stability of the integrated reaction zone by providing a uniform temperature profile and strategic temperature control.

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## **“APPARATUS FOR PRODUCING HYDROGEN”**

### **BACKGROUND OF THE INVENTION**

5 This invention relates to reactors for the production of hydrogen used in conjunction with a fuel cell for the production of electricity. More particularly, the invention relates to an integrated reactor for converting a hydrocarbon or alcohol to produce a fuel stream for an electrochemical reaction zone.

10 Systems for contacting solids and particulate material are well known and routinely employed in the processing of gases, the production of chemicals, and the refining of petroleum. The particulate materials in most cases comprise catalysts or adsorbents and the process streams are gaseous or liquid mixtures of reactants, product or streams undergoing separation.

15 One particularly well-known method of contacting particulate material with a fluid stream retains the particulate solid material as a bed of particulate material through which the fluid stream passes. A multitude of arrangements with various bed geometries are known for contacting the particulate material with the fluid streams. Such arrangements include radial flow beds where particulate solids are retained in an annular ring or downflow or upflow beds where fluid streams pass through a cylindrical bed or laminar bed of particulate solids.

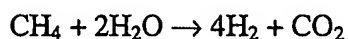
20 The use of fuel cells to generate electrical power for electricity or to drive a transportation vehicle relies upon the generation of hydrogen. Because hydrogen is difficult to store and distribute, and because hydrogen has a low volumetric energy density compared to fuels such as gasoline, hydrogen for use in fuel cells will have to be produced at a point near the fuel cell, rather than be produced in a centralized refining facility and distributed like gasoline. To be effective, hydrogen generation for fuel cells  
25 must be smaller, simpler, and less costly than hydrogen plants for the generation of industrial gasses. Furthermore, hydrogen generators for use with fuel cells will have to be integrated with the operation of the fuel cell and be sufficiently flexible enough to efficiently provide a varying amount of hydrogen as demand for electric power from the fuel cell varies.

Hydrogen is widely produced for chemical and industrial purposes by converting materials such as hydrocarbons and methanol in a reforming process to produce a synthesis gas. Such production usually takes place in large facilities which are rarely turned down in production for even a few days per year. In addition, the operation of the industrial hydrogen production facilities is often integrated with associated facilities to improve the use of energy for the overall complex. Synthesis gas is the name generally given to a gaseous mixture principally comprising carbon monoxide and hydrogen, but also possibly containing carbon dioxide and minor amounts of methane and nitrogen. It is used, or is potentially useful, as feedstock in a variety of large-scale chemical processes, for example: the production of methanol, the production of gasoline boiling range hydrocarbons by the Fischer-Tropsch process and the production of ammonia.

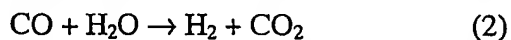
Processes for the production of synthesis gas are well known and generally comprise steam reforming, autothermal reforming, non-catalytic partial oxidation of light hydrocarbons or non-catalytic partial oxidation of any hydrocarbons. Of these methods, steam reforming is generally used to produce synthesis gas for conversion into ammonia or methanol. In such a process, molecules of hydrocarbons are broken down to produce a hydrogen-rich gas stream. A paper titled "Will Developing Countries Spur Fuel Cell Surge?" by Rajindar Singh, which appeared in the March 1999 issue of *Chemical Engineering Progress*, page 59-66, presents a discussion of the developments of the fuel cell and methods for producing hydrogen for use with fuel cells. The article particularly points out that the partial oxidation process is a fast process permitting small reactors, fast startup, and rapid response to changes in the load, while steam reforming is a slow process requiring a large reactor and long response times, but operates at a high thermal efficiency. The article highlights one hybrid process which combines partial oxidation and steam reforming in a single reaction zone as disclosed in US-A-4,522,894.

Modifications of the simple steam reforming processes have been proposed to improve the operation of the steam reforming process. In particular, there have been suggestions for improving the energy efficiency of such processes in which the heat available from the products of a secondary reforming step is utilized for other purposes within the synthesis gas production process. For example, processes are described in US-A-4,479,925 in which heat from the products of a secondary reformer is used to provide heat to a primary reformer.

The reforming reaction is expressed by the following formula:



where the reaction in the reformer and the reaction in the shift converter are respectively expressed by the following simplified formulae (1) and (2):



In the water gas shift converter which typically follows a reforming step, formula (2) is representative of the major reaction.

US-A-4,925,456 discloses a process and an apparatus for the production of  
10 synthesis gas which employs a plurality of double pipe heat exchangers for primary reforming in a combined primary and secondary reforming process. The primary reforming zone comprises at least one double-pipe heat exchanger-reactor and the primary reforming catalyst is positioned either in the central core or in the annulus thereof. The invention is further characterized in that the secondary reformer effluent is  
15 passed through which ever of the central core or the annulus is not containing the primary reforming catalyst counter-currently to the hydrocarbon-containing gas stream.

US-A-5,181,937 discloses a system for steam reforming of hydrocarbons into a hydrogen rich gas which comprises a convective reformer device. The convective reformer device comprises an outer shell enclosure for conveying a heating fluid  
20 uniformly to and from a core assembly within the outer shell. The core assembly consists of a multiplicity of tubular conduits containing a solid catalyst for contacting a feed mixture open to the path of the feed mixture flow such that the path of the feed mixture flow is separated from the heating fluid flow in the outer shell. In the process, an autothermal reformer fully reforms the partially reformed (primary reformer) effluent  
25 from the core assembly and supplies heat to the core assembly by passing the fully reformed effluent through the outer shell of the convective reforming device.

WO 99/36351 discloses a reformer reactor for producing a hydrogen-rich gas which includes 4 sequentially adjacent reaction zones and a product gas collection space. The reformer reactor comprises a reactor geometry such that the flow path is directed in  
30 diverging radial directions away from the first reaction zone and through the subsequent zones. In addition the reactor is provided with a heat exchange means disposed in one of

the downstream reaction zones to use heat developed in the reaction zones to preheat the feed stream. This heat exchange means is employed to regulate the heat exchange in the reactor to achieve a desired temperature in the catalyst zone. The heat exchange means disclosed in WO 99/36351 comprises helical tube sections disposed in the catalyst zones  
5 to the feed stream.

WO 99/36351 and similar axial flow arrangements based on locating adjacent reaction zones in a manner to facilitate heat exchange and then disposing within those reaction zones coils of heat exchanger tubes suffer from the practical problem of regulating and controlling the heat exchange in the coils and the mechanical stresses  
10 generated in the heat exchange tubes. Simple axial flow systems employing double pipe heat exchange zones which rely on simple convection between adjacent zones are not able to remove a sufficient amount of heat because the heat exchange medium is typically limited to the gaseous reaction mixture which has a low thermal conductivity. In addition, the placement of coils within relatively narrow catalytic zones of these close-  
15 coupled reaction systems can result in poor flow distribution of reactants and an unfavorable temperature profile in the direction perpendicular to the flow of the reactants. The equilibrium reactions presented herein above are sensitive to temperature and may exhibit a reversal when the temperature of the particular reaction zone falls outside an effective operating range. Furthermore, two phase flow conditions may arise  
20 within the coils resulting in vibration and bumping which can result in mechanical failure of the coil and can raise safety issues related to hydrogen leaks and fire. Integrated reaction systems are sought which avoid the above-mentioned problems.

It is an objective of the present invention to provide compact fuel processor for use with fuel cell systems.

25 It is an objective of the present invention to provide a fuel processor reactor which provides a uniform temperature profile to avoid wide temperature swings and minimize mechanical and thermal stress.

It is an objective of the present invention to provide an isothermal heat medium having a heat capacity greater than the feed or the reactor products to strategically  
30 control the temperature throughout the fuel processor zone.

### SUMMARY OF THE INVENTION

The present invention is directed to a fuel processor to be used in conjunction with a fuel cell for the generation of electric power. The reactor portion of the fuel processor comprises an arrangement wherein particulate solids are disposed in concentric annular catalyst zones and wherein the catalyst is retained in relatively narrow vertically extended annular flow channels within the concentric annular catalyst zones. The particulate solids remain in a fixed position and the fluids move in a direction transverse to the vertical direction. Screens or monolithic catalytic members are used to define the annular catalyst zones. Strategically positioned tubes are used to define vertical flow channels for heat transfer fluids. This is particularly important in applications that require or benefit from heating or cooling of the particulate solids and fluids within the reaction zones to control the temperature of a reaction or other processing. In such arrangements, the vertical tubes provide a large area of heat transfer surface by which a heat transfer fluid may indirectly contact one surface of the tube while the other surface contacts the particulate solids and fluids. For example, indirect heat transfer in which a heat exchange fluid contacts a catalyst and a reaction fluid or a reaction fluid in combination with another fluid can be used to supply or withdraw the heat of reaction in an endothermic or exothermic process to establish isothermal conditions in the reaction zone.

In one embodiment, the present invention comprises an apparatus for generating hydrogen from a fuel stream for use in conjunction with a fuel cell. The apparatus comprises a vertically aligned reforming reaction zone comprising an oxidation zone disposed vertically over a reaction zone base and a flow transition zone to collect an effluent of the oxidation zone. The flow transition zone is established by sealingly disposing a reforming catalyst zone which is radially permeable and surrounds the oxidation zone on the reaction zone base between the reaction zone base and an insulated reaction zone ceiling at an effective distance from the oxidation zone. A plurality of radially permeable reaction zones surrounds the vertically aligned reforming reaction zone and is disposed sealingly between the reaction zone base and the insulated reaction zone ceiling. A radial product collection zone is disposed sealingly about an outer catalyst zone of the plurality of radially permeable catalyst zones. A plurality of vertically aligned heat exchanger tubes is disposed in at least one of the radially permeable reaction zones and extends through the reactor zone base and the insulated reaction zone ceiling. The plurality of vertically aligned heat exchanger tubes is in fluid

communication with an essentially isothermal fluid, wherein the vertically aligned reforming reaction zone is at least partially submerged in the essentially isothermal fluid.

In a further embodiment, the present invention is an apparatus for generating hydrogen from a fuel stream for use in conjunction with a fuel cell. The apparatus comprises a reaction zone comprising a top plate and a reactor base. The reactor base has a top face, a bottom face, and a vertical axis extending therethrough. A hollow cylinder oriented vertically and rigidly is disposed on the top face of the reactor base about the vertical axis and extends upwardly toward the top plate, forming a first catalyst zone. The first catalyst zone contains an inert catalyst layer at the top face and a partial oxidation catalyst layer above the inert catalyst layer. The first catalyst zone is in fluid communication with a feed inlet which extends downwardly through the circular base. A second catalyst zone surrounds the first catalyst zone and extends between the base and the top plate and is sufficiently offset from the first catalyst zone to establish a flow transition zone from vertical to radial feed flow. The second catalyst zone contains a reforming catalyst. A third catalyst zone is disposed on and surrounds the second catalyst zone and extends between the reactor base and the top plate. The third catalyst zone contains a high-temperature shift catalyst. A fourth catalyst zone is disposed on and surrounds the third catalyst zone and extends between the reactor base and the top plate. The fourth catalyst zone contains a low-temperature water gas shift catalyst. A plurality of vertical heat exchanger tubes extends between the reactor base and the top plate. The plurality of vertical heat exchanger tubes is sealingly disposed on and extends through the top plate and the reactor base. A layer of insulation is disposed over the top plate and below the bottom face of the reactor base and extends outwardly from the vertical axis toward the fourth catalyst zone. The reaction zone is rigidly disposed on supporting legs in a boiler vessel which has an interior space and is at least partially filled with a fluid. The vertical heat exchanger tubes are in fluid communication with the fluid, and the reaction zone is at least partially submerged in the fluid to provide cooling for the fourth reaction zone and for the generation of steam through the thermosiphon action of boiling fluid in the vertical heat exchange tubes.

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### **BRIEF SUMMARY OF THE DRAWINGS**

FIG. 1 provides a cross-sectional view of the present invention.

FIG. 2 shows a cut-away perspective of the present invention.



### **DETAILED DESCRIPTION OF THE INVENTION**

This invention applies to arrangements for the integration of highly exothermic and endothermic reaction zones into a single reaction zone with the ability to manage the heating and cooling requirements in a manner to provide a stable operation of the combination. The key to achieving a stable operation is the attainment of an essentially isothermal heat sink which provides a continuous source of cooling to a highly exothermic reaction system while providing a continuous source of steam at a critical point in the process. FIG. 1 shows a fuel processor/boiler 100 of the present invention. The feed stream to the fuel processor/boiler 100 comprises a hydrocarbon such as natural gas or an oxygenate such as methanol, ethanol, or propanol and steam. Hydrocarbon feeds can also include LPG, liquefied petroleum gas, gasoline, or diesel, and oxygenates can include the general class of alcohols, ethers, and ketones. The feed stream may be a simple admixture of the hydrocarbon or oxygenate and steam, or the feed stream may be pre-processed in a pre-reforming zone at moderate reforming conditions over a reforming catalyst in the presence of steam at an effective steam-to-carbon ratio to at least partially convert the hydrocarbon or oxygenate to produce carbon oxides, hydrogen and water. The feed stream comprising a hydrocarbon or an oxygenate and water is passed to the fuel processor/boiler 100 in conduit 10 which enters the bottom of the thermosiphon boiler vessel 58 and extends through a layer of insulation 22 below the reactor base 11 of the vertically aligned reforming zone 60, or integrated reforming zone, and into an inert catalyst zone 12, surrounded by an inner wall forming the inert catalyst zone, a partial oxidation zone, and extending above the partial oxidation zone to form a partial oxidation zone outlet 16. A layer of partial oxidation catalyst is disposed over the inert catalyst in a partial oxidation zone 14. In the partial oxidation zone 14 the feed stream is contacted with the partial oxidation catalyst in the presence of an oxygen-containing stream to produce hydrogen, carbon oxides, and water in an exothermic reaction. The heat released in the partial oxidation reaction provides an oxidation effluent stream which is withdrawn from the partial oxidation zone outlet 16 into a flow transition zone 18 wherein the direction of the flow of the oxidation effluent stream is redirected from axial flow to radial flow. The flow transition zone 18 is bounded radially by a reforming zone 24, spaced an effective distance away from the partial oxidation zone 14 to permit the flow transition to radial flow to take place. The flow transition zone 18 is established by the space between the partial oxidation zone, the reaction zone

ceiling 13. The reaction zone ceiling is insulated with a layer of insulation 20 which covers the partial oxidation zone 14 and extends over the reforming zone 24 to retain the heat generated in the partial oxidation zone for use in the reforming reaction which is endothermic. It is particularly important to insulate both the flow transition zone 18 and the reforming zone 24 to prevent the cooling of the reforming catalyst at the boundaries defined by the reactor base 11 and the reaction zone ceiling 13. Thus, the partial oxidation zone 14 is sealingly surrounded by the radially permeable reforming zone 24 such that the effluent from the partial oxidation zone 14 passes radially through the reforming zone 24. A plurality of reaction zones surround the reforming zone 24 in a close-coupled manner and extending between the reactor base 11 and the reaction zone ceiling 13. The plurality of close-coupled reaction zones, which may be monoliths containing catalysts or fixed beds separated and supported by screens or other rigid permeable boundaries 15 are selected from the group consisting of a high-temperature water gas shift zone 26, a low-temperature water gas shift zone 28, and a preferential oxidation zone (not shown), and combinations thereof. Returning to FIG. 1, a high-temperature water gas shift zone 26 surrounds the reforming zone 24. In the high-temperature water gas shift reaction zone 26, at least a portion of the carbon monoxide produced in the reforming and oxidation reactions is converted to additional amounts of hydrogen by combination with water. The effluent from the high-temperature shift reaction zone 26 continues flowing radially into a low-temperature water gas shift zone 28. It is critical that the temperature in the vicinity of the transition between the high-temperature water gas shift zone and the low-temperature water gas shift zone be controlled. According to the present invention, a plurality of vertically aligned heat exchanger channels or pipes 32 are disposed in the low-temperature water gas shift zone to provide cooling of the low-temperature water gas shift zone and to the high-temperature water gas shift zone. The vertically aligned heat exchanger tubes provide a first plurality of thermosiphon boiler tubes. Preferably, the first plurality of thermosiphon tubes 32 are located at a point between about  $\frac{1}{2}$  and about  $\frac{3}{4}$  of the radial distance into the low-temperature water gas shift zone 28. By locating the first plurality of thermosiphon tubes at this point in the low-temperature water gas shift zone 28, a portion of the cooling of the high-temperature water gas shift zone takes place as well. Should additional cooling of the high-temperature water gas shift zone 26 be required, a second plurality of thermosiphon tubes 34 may be disposed at any point between  $\frac{1}{4}$  of the depth

of either the high-temperature water gas shift zone and  $\frac{1}{4}$  the depth of the low-temperature water gas shift zone. The advantage of the use of the thermosiphon tubes in the high and low-temperature water gas shift zones 26 and 28 is that the temperature of the thermosiphon tubes is essentially a constant value approximately equal to the boiling point of water at the pressure of the steam being generated from the thermosiphon boiler vessel 58. A product gas collection zone 30 sealingly surrounds the last annual reaction zone in the plurality of close-coupled reaction zones. A product gas conduit 50 provides for the removal of the product gas from the integrated reaction zone 60. The integrated reaction zone 60 is at least partially submerged in a layer of fluid such as water in a thermosiphon boiler vessel 58. The plurality of thermosiphon tubes 32 and 34 are in fluid communication with the fluid in the boiler vessel. The level of the layer fluid with respect to the integrated reaction zone 60 is maintained by a standpipe 36 which is disposed inside the boiler vessel and is supported by a plurality of standpipe supports 42 rigidly disposed inside the thermosiphon boiler vessel 58. The integrated reaction zone 60 is supported by a plurality of legs 38 disposed inside the thermosiphon boiler vessel 58 and rigidly attached to the reactor base 11 and insulation layer 22 of the integrated reaction zone 60. The boiler vessel 58 is provided with a steam outlet 52 at the top of the boiler vessel and a water inlet 40 below the level of the fluid 54. The fluid in the thermosiphon boiler vessel can comprise a high boiling additive such as a glycol such as ethylene glycol and/or a glycol ether to prevent the fluid from freezing. When the thermosiphon boiler is generating steam, the high boiling additive remains within the thermosiphon boiler vessel 58 by being less volatile than the water. In one embodiment, steam generated in the boiler is conveyed to a steam tank (not shown) at a predetermined height above the boiler vessel by a conduit between the steam outlet and the steam tank and a water conduit is provided to return at least a portion of any condensed fluid and a water conduit is provided to return at least a portion of the condensed fluid to the boiler. Any make-up water is introduced by a make-up conduit to the boiler vessel 58. The predetermined height of the steam tank above the boiler vessel is based on the effective height to establish a thermosiphon effect in the boiler vessel 58 and is well known to those skilled in the art. If required, additional heating can be provided by passing burner exhaust gas through combustion gas conduit 46 through a fluid heat exchanger 44 and withdrawing a cooled combustion effluent stream from conduit 48. Alternatively, the

thermosiphon flow in the plurality of thermosiphon tubes can be established as described in FIG. 2 in connection with the use of a standpipe.

Referring to FIG. 2, a cross-sectional perspective view of the integrated reaction zone 160 is shown vertically aligned in a boiler vessel 200. The feed stream comprising fuel and steam is introduced axially through a conduit 110 from below the base 111 into an inert catalyst zone 112. A partial oxidation zone 114 containing a partial oxidation catalyst is disposed above the inert catalyst zone 112. The effluent from the partial oxidation zone 114 is withdrawn from the partial oxidation zone outlet 116 into a flow transition zone 118 between the partial oxidation zone outlet and a reforming zone 124, or the first catalyst zone of a plurality of radially permeable catalyst zones surrounding the partial oxidation zone 114. Each of the plurality of radially permeable catalyst zones is sealingly disposed on the base 111 and an insulated reaction zone ceiling (not shown in this view). The plurality of catalyst zones includes a reforming zone, a high-temperature water gas shift zone, a low-temperature water gas shift zone, a preferential oxidation zone, and combinations thereof. The last of the plurality of radially permeable catalyst zones is surrounded sealingly by a product gas collection zone 130 to collect the radially transmitted product gas and withdraw the product gas via conduit 150. The product gases collected in this manner may be passed to a fuel cell or a preferential oxidation zone for the further reduction of carbon monoxide to less than about 50 ppm-vol. In order to control the distribution of heat within the integrated reaction zone 160, a first plurality of vertically aligned thermosiphon tubes 132 is disposed in the low-temperature water gas shift zone 128. The vertically aligned thermosiphon tubes are in fluid communication with a fluid in the boiler vessel 200. Fluid level in the boiler vessel 200 is maintained with a standpipe 136 such that the fluid at least partially covers the integrated reaction zone 160, and boiling takes place within the vertically aligned thermosiphon tubes 132. The boiling of the fluid within the thermosiphon tubes provides an essentially isothermal condition in the integrated reaction zone 160 and establishes a thermosiphon flow of steam, boiling water, or a mixture of steam and water at its boiling point. If required for additional heat removal, a second plurality of thermosiphon tubes 134 is disposed either in the high-temperature water gas shift zone 126 or a portion of the low-temperature water gas shift zone 128 adjacent to the high-temperature water gas shift zone 126. On start-up, a combustion gas heat exchanger 144 is provided to employ combustion gas in conduit 146 to heat the fluid in the boiler vessel 200. The cooled

combustion gas is removed in conduit 148. A steam outlet conduit 152 to collect the steam produced in the thermosiphon tubes 134 and 132 is provided at the top of the boiler vessel and a water inlet conduit is provided to permit water to be returned from the thermosiphon tank (if present), or to provide for make-up water. Thermosiphon flow can  
5 be established within the plurality of thermosiphon tubes 132 and 134 without the use of an external collection tank by establishing a fluid level in the boiler vessel at an effective height. The effective height of the fluid level in the boiler is maintained by a standpipe of sufficient height above the integrated reaction zone 160 and extending below the integrated reaction zone 160. Additional heat to provide additional steam to obtain a  
10 desired steam-to-carbon ratio in the reforming zone for feed streams heavier than methane can be obtained by heat exchange with combustion gases in heat exchanger 144 which is disposed in the fluid.

In all cases, it is understood that the above-identified arrangements are merely illustrative of the many possible specific embodiments which represent application of the  
15 present invention. Numerous and varied other arrangements can readily be devised in accordance with the principles of the present invention without departing from the spirit and scope of the invention.

**CLAIMS:**

1. An apparatus for generating hydrogen from a fuel stream for use in conjunction with a fuel cell, said apparatus comprising:

a vertically aligned reforming reaction zone (60) comprising an oxidation zone (14) disposed vertically over a reaction zone base (11),

a flow transition zone (18) to collect an effluent of the oxidation zone (14), said flow transition zone (18) established by sealingly disposing a reforming catalyst zone (24) being radially permeable and surrounding the oxidation zone (14) on the reaction zone base (11) between the reaction zone base (11) and an insulated reaction zone ceiling (13) at an effective distance from the oxidation zone (14);

a plurality of radially permeable reaction zones (24, 26, 28) surrounding the vertically aligned reforming reaction zone (60) disposed sealingly between the reaction zone base (11) and the insulated reaction zone ceiling (13);

a radial product collection zone (30) disposed sealingly about an outer catalyst zone (28) of the plurality of radially permeable catalyst zones (24, 26, 28); and

a plurality of vertically aligned heat exchanger tubes (32) disposed in at least one of the radially permeable reaction zones (24, 26, 28) and extending through the reactor zone base (11) and the insulated reaction zone ceiling (13), said plurality of vertically aligned heat exchanger tubes (32) being in fluid communication with an essentially isothermal fluid (54);

wherein the vertically aligned reforming reaction zone (60) is at least partially submerged in the essentially isothermal fluid (54).

2. The apparatus of claim 1 wherein the plurality of radially permeable reaction zones is selected from the group consisting of a high-temperature shift zone (26), a low-temperature gas shift zone (28), a preferential oxidation zone and combinations thereof.

3. The apparatus of claim 1 further comprising a conduit (50) for withdrawing a product gas stream from the radial product collection zone (30).

4. The apparatus of claim 1 wherein the essentially isothermal fluid comprises boiling water or a mixture of steam and water at its boiling point.

5. The apparatus of claim 1 further comprising a boiler vessel (100) having a hollow interior, a steam outlet conduit (52), and a water inlet conduit (40) wherein the vertically aligned reforming reaction zone (60) is rigidly disposed in the hollow interior of the boiler vessel (100).

5        6. The apparatus of claim 5 further comprising a conduit for connecting the steam outlet (52) to an overhead collection tank having an upper zone and a lower zone, said upper zone having a steam distribution conduit and said lower zone having a water make-up conduit and a water return conduit connecting the overhead collection tank with the water inlet conduit.

10       7. The apparatus of claim 1 further comprising an inert catalyst zone (12) disposed between the oxidation zone (14) and the reaction zone base (11).

8. The apparatus of claim 6 further comprising a standpipe (36) vertically aligned and disposed in the interior of the boiler (100) to maintain a level of isothermal fluid (54) in the interior of the boiler at least partially covering the vertically aligned  
15 reaction zone (60).

9. The apparatus of claims 1-8 wherein the apparatus further comprises

a reaction zone comprising a top plate (13) and a reactor (11), said reactor base (11) having a top face, a bottom face, and a vertical axis extending therethrough;

20       a hollow cylinder (116) oriented vertically and rigidly disposed on the top face of the reactor base (11) about said vertical axis and extending upwardly toward the top plate (13), forms the vertical catalyst zone, containing the inert catalyst layer (12) at the top face and the partial oxidation catalyst layer (14) above the inert catalyst layer (12), and the vertical catalyst zone being in  
25 communication with a feed inlet extending downwardly through the reactor base;

a first catalyst zone (24) of the radially permeable catalyst zone (24, 26, 28) contains the reforming catalyst and surrounds the vertical catalyst zone and extends between the reactor base (11) and the top plate (13) and sufficiently offset from the first catalyst zone to establish the flow transition zone (18) from  
30 vertical to radial feed flow to said second catalyst zone;

a second catalyst zone (26) disposed on and surrounding the first catalyst zone extends between the reactor base (11) and the top plate (13), said second catalyst zone contains the high-temperature shift catalyst;

a third catalyst zone (28) disposed on and surrounding the second catalyst zone (28) and extends between the reactor base (11) and the top plate (13), said third catalyst zone containing the low-temperature water gas shift catalyst and the plurality of vertical heat exchanger tubes (32) extending between the reactor base (11) and the top plate (13), said plurality of vertical heat exchanger tubes (32) being sealingly disposed on and extending through the top plate (13) and the reactor base (11); and

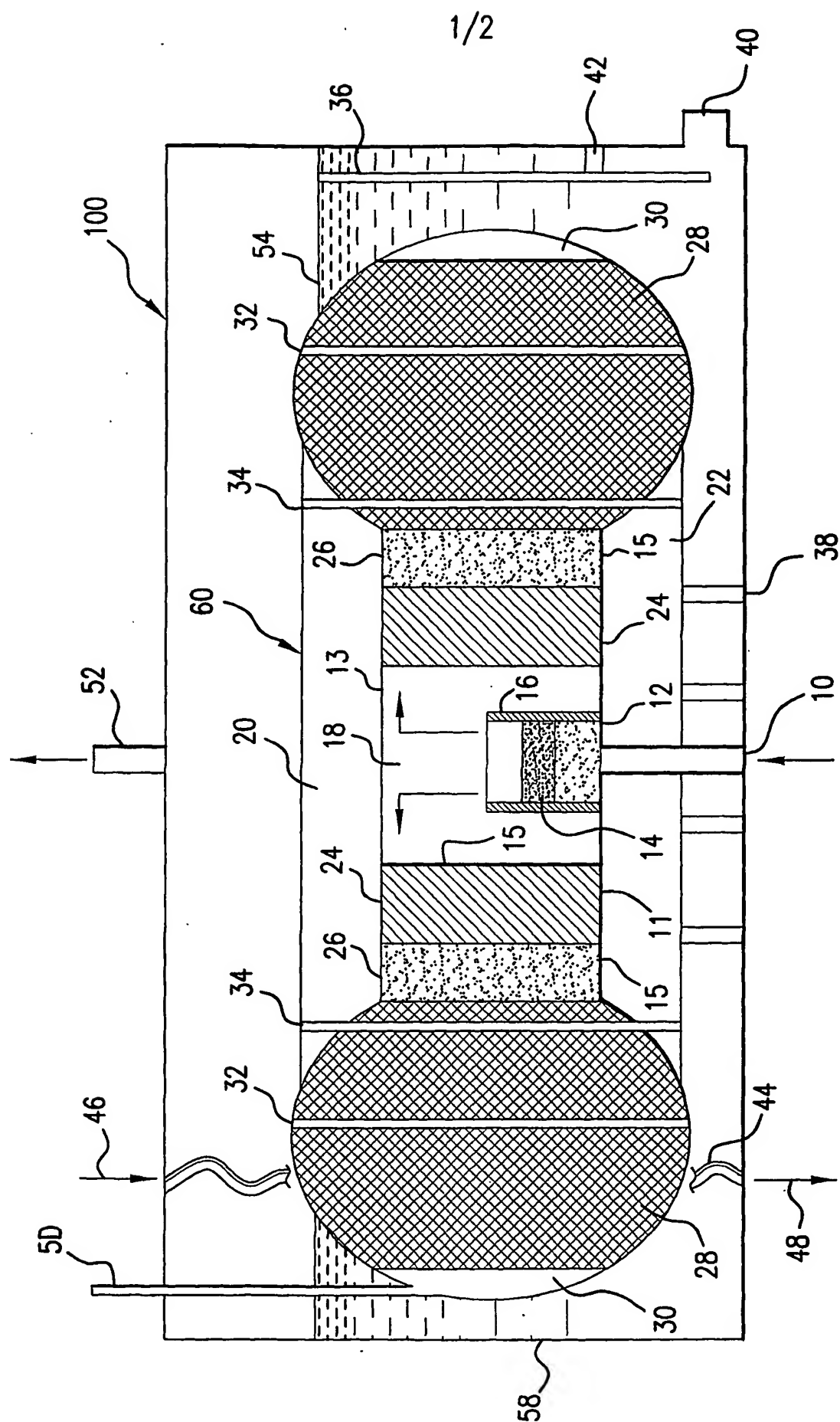
a layer of insulation (20, 22) disposed over the top plate (13) and below the bottom face of the reactor base and extending outwardly from the vertical axis toward the third catalyst zone; and

the reaction zone is disposed in a boiler vessel (100) having an interior space at least partially filled with the essentially isothermal fluid wherein the vertical heat exchanger tubes (32) are in fluid communication with the fluid and the reaction zone (60) is at least partially submerged in the fluid to provide cooling for the third reaction zone and for the generation of steam through thermosiphon action of boiling fluid in said vertical heat exchanger tubes (32).

10. The apparatus of claim 9 further comprising a second plurality of vertical heat exchanger tubes (32) disposed in the third catalyst zone (28) extending through the reactor top plate (13) and the reactor base (11) and being in fluid communication with the fluid in the interior space of the thermosiphon boiler.

11. The apparatus of claim 9 wherein the first, second and third, catalyst zones (24, 26, 28) comprise catalyst monoliths.





**FIG. 1**

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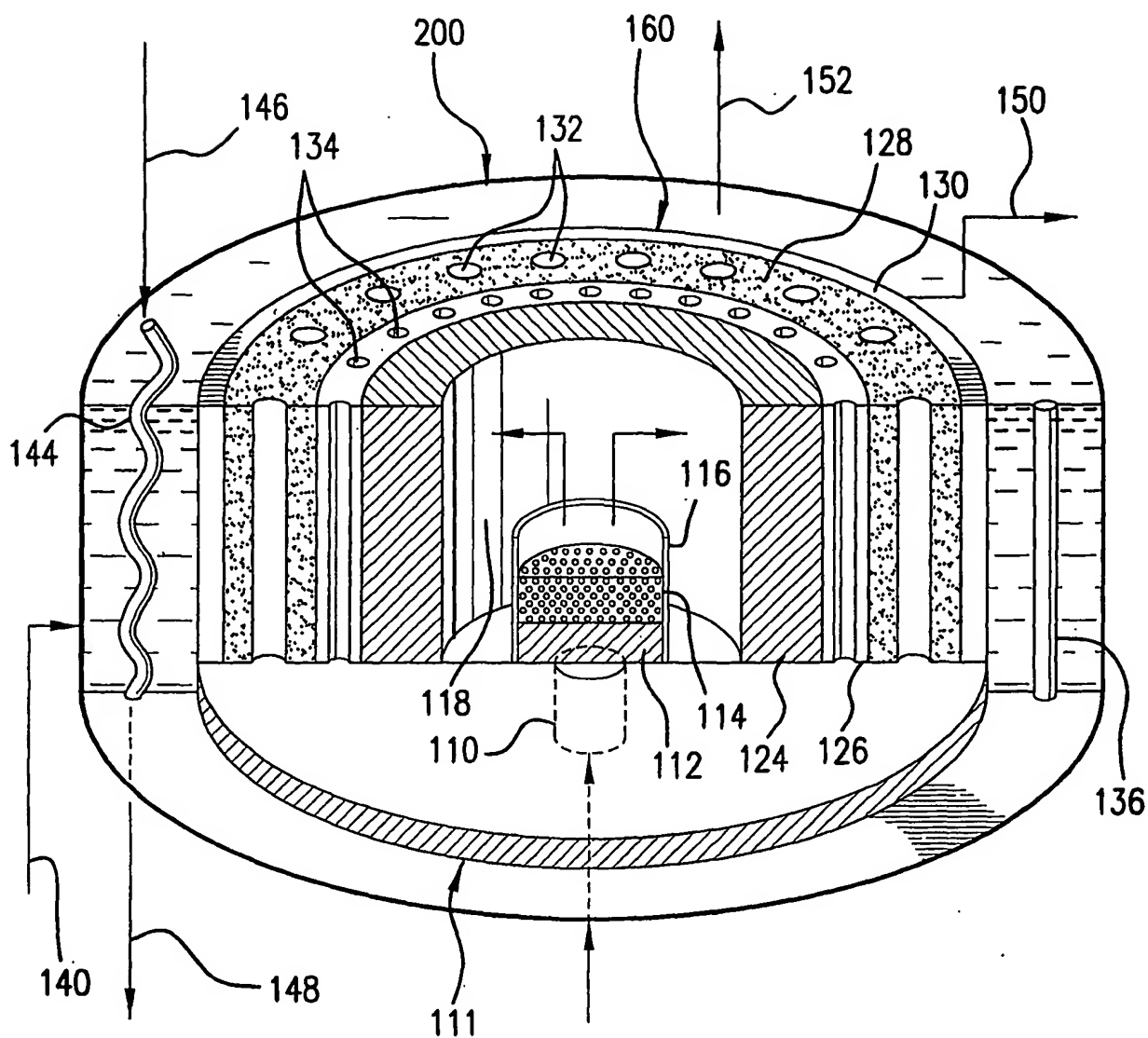


FIG.2

## A. CLASSIFICATION OF SUBJECT MATTER

IPC 7 C01B3/38 C01B3/48 B01J8/04 C01B3/32 C01B3/58

According to International Patent Classification (IPC) or to both national classification and IPC

## B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

IPC 7 C01B B01J

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practical, search terms used)

EPO-Internal, WPI Data, PAJ

## C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
A	US 3 541 729 A (DANTOWITZ PHILIP) 24 November 1970 (1970-11-24) column 7, line 6-27; claim 1; figure 1 ---	1-9
A	US 5 006 316 A (ZARDI UMBERTO ET AL) 9 April 1991 (1991-04-09) the whole document ---	1-9
A	EP 0 600 621 A (ROLLS ROYCE & ASS) 8 June 1994 (1994-06-08) claim 1; figures 1-3 ---	1-9
A	WO 99 36351 A (LITTLE INC A) 22 July 1999 (1999-07-22) claims 1-7, 17-22; figures 1-7 -----	1-9



Further documents are listed in the continuation of box C.



Patent family members are listed in annex.

## \* Special categories of cited documents:

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Date of the actual completion of the international search

18 October 2001

Date of mailing of the international search report

25/10/2001

Name and mailing address of the ISA

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